Ab-initio Simulations of the Nonequilibrium Properties of Warm Dense Gold

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Recent experiments on gold thin films suggested that electrical and optical properties of metals in the warm dense matter regime can be measured by performing time-resolved measurements after the illumination of a metallic thin film by a short-pulse laser [1]. In this experimental configuration, the laser energy is uniformly deposited on a gold thin film with intensity ranging from 0.5 x 10^6 to 2 x 10^7 J/Kg. As the laser energy is directly absorbed by the conduction band electrons, the system is out of equilibrium with different electron and ion temperatures immediately after illumination. Optical properties are subsequently measured while the ions and electrons relax to their new equilibrium temperatures.

Molecular dynamics (MD) simulations based on density functional have been rather successful at describing the physical properties of equilibrium states reached in shock experiments. However, the nonequilibrium situation created in this experimental setup poses new challenges as the simulation method needs to accurately describe the time evolution of the sample after illumination by the short-pulse laser. The subsequent transfer of energy between the electrons and ions is a nonadiabatic effect, and its formal treatment is beyond the current *ab-initio* capabilities. We follow previous work by Ivanov et al. [1] and introduce the electron-phonon coupling by using a thermostat in the *ab-initio* method to produce time resolved optical conductivities for this nonequilibrium simulation.

As *ab-initio* simulations are limited by the size of the simulation cell that can be used, we further used a hybrid MD method, as proposed by Ivanov et al., to characterize the time evolution of the lattice structure as a function of laser energy. As illustrated in Fig. 1, the hybrid MD approach couples a continuum model, the two temperature model, with a classical MD method to follow the evolution of both the electron and ion temperatures as a function of time. Using this method, we were able to calculate the time needed for the fcc lattice to melt as a function of laser energies and to identify a quasi-steady state observed in the experimental optical conductivities as the duration of a superheated fcc state.

Using this result, we performed *ab-initio* calculations of the optical properties assuming that the underlying ion lattice preserves its initial fcc structure along

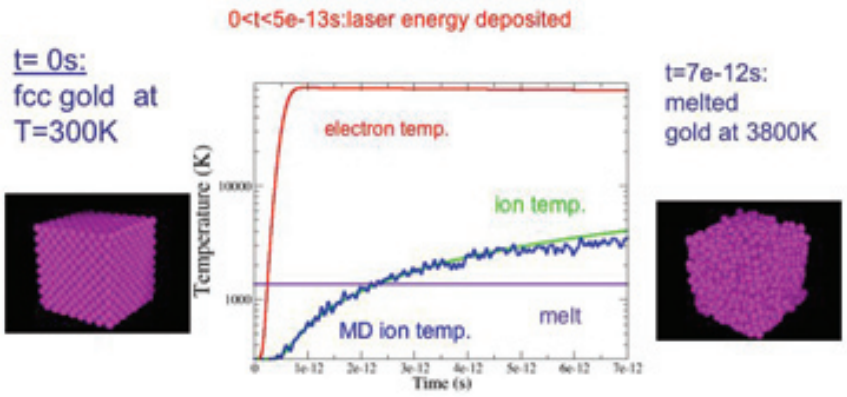

Fig. 1.
Variation of the electronic and ionic temperatures as given by the TTM.

the quasi-steady state during which experimental optical properties are averaged. In Fig. 2, we compare the variation of the real and imaginary parts of the optical conductivity as a function of laser energy and at the frequency measured experimentally. We find a remarkable agreement with the experimental data, especially for the real part of the conductivity. Overall, the calculations reproduce both the magnitude and the trend as a function of laser energy observed experimentally. This clearly shows that the experimental measurements are compatible with the existence of a metastable fcc state lasting for the duration of the quasi-steady state. Finally, we further tested this assumption by calculating conductivities of a liquid state equilibrated at 2000 K. In this situation, the dependence on the laser energy was obtained by calculating conductivities using electron temperatures given by the TTM model. We see, in Fig. 2, that for both the real and imaginary parts, the transition to a liquid state should be clearly visible in measuring the optical properties and at all the laser energies studied here.

In conclusion, we find, using a combination of classical and *ab-initio* simulations, that for the conditions created experimentally, the ions mostly preserved their initial fcc structure for several picoseconds before evolving into a liquid state. The near perfect agreement with the experimental measurements on gold thin films first showed that the quasi-steady state observed experimentally corresponds to a metastable fcc state where the lattice has not yet reacted to the elevation of the electron temperature. This study also demonstrates that *ab-initio* simulations combined with linear response theory provide a powerful tool to calculate the physical properties of nonequilibrium states generated by laser-matter interactions or short-pulse high energy-density physics experiments [3].

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[2] K. Widmann, et al., *Phys. Rev. Lett.* **92**, 125002 (2004).
[3] S. Mazevet, et al., *Phys. Rev. Lett.* **95**, 085002 (2005).

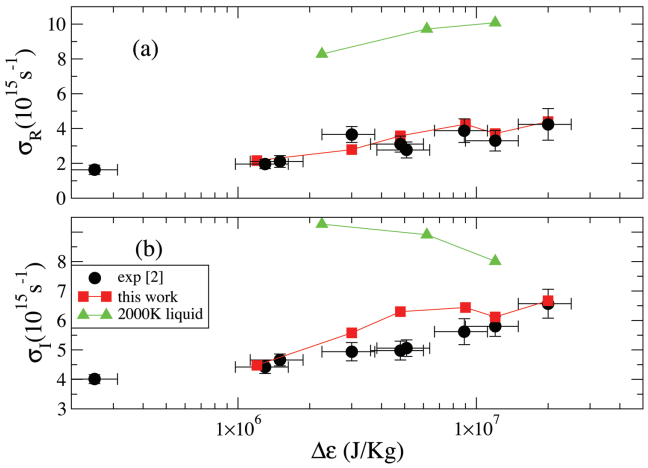

Fig. 2.
Comparison of the variation of the real and imaginary parts of the optical conductivity as a function of laser energy and at the frequency measured experimentally.